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Three-Phase Combustion Modeling: Frozen Ozone, a Prototype System

by Martin S. Miller

ARL-TR-1320

March 1997

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ARL-TR-1320

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Three-Phase Combustion Modeling: Frozen Ozone, a Prototype System

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Abstract

A number of efforts are currently underway in the United States to model the self-sustained combustion of solid energetic materials at the level of fundamental physical processes and elementary chemical reactions. Since many of these materials burn, at least at some pressures, with a liquid surface layer, these models must address phenomena in three physical phases. Most of these modeling efforts have focused on pure RDX as a prototype. From a systems viewpoint, this was a natural choice, since it, or materials like it, is one of the components of modern propellants. It is argued here that, from a scientific viewpoint, RDX is already too complex with too many uncertain mechanisms and unavailable supporting data to serve this role effectively. We discuss here a number of issues related to the condensed phase and interfacial phenomena which have not been previously identified and which warrant more detailed research. In this report, frozen ozone is adopted as a more suitable prototypical three-phase system, and progress toward addressing this system is presented. The first new process modeled was a gas/surface reaction. A detailed analysis was performed on the reaction mechanisms at play in the ozone flame and how they are affected by the heterogeneous reaction. Other new mechanisms are associated with the multicomponent nature of the liquid surface layer and will be addressed in future work.

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1. INTRODUCTION

Over the last decade there has been an increasing interest and optimism that one might be able to describe the fundamental chemical and physical processes of energetic-material combustion in an *a priori* mathematical model. Nitramine combustion has been the subject of the bulk of this activity. One of the first of these models was proposed by Ben-Reuven, et al.^{1,2,3} who, though treating the gas-phase chemistry as two global steps and the liquid chemistry as one global step, nevertheless considered all three phases, coupling the mass, species, and energy conservation equations using appropriate boundary conditions at the phase interfaces (including phase-transition enthalpies). Little decomposition of either RDX (cyclotrimethylenetrinitramine) or HMX (cyclotetramethylenetetranitramine) was found to occur in the condensed phase, so that the dominant surface regression mechanism was nitramine evaporation at the surface. Later, Williams and coworkers^{4,5}, developed RDX- and HMX-combustion models, which considered a single global reaction in each of the liquid and gas phases but also allowed for nitramine vaporization at the surface. While agreeing with Ben-Reuven, et al. that little decomposition occurs in the liquid phase for RDX, they found that 30% or more of HMX decomposed in the condensed phase. They did not treat the liquid layer explicitly but did consider the effects of two-phase flow in the condensed phase. Next, Melius⁶ ratcheted up the complexity in his RDX combustion model by considering 38 gas-phase species related by 158 reactions, one global condensed-phase reaction, and evaporation of RDX at the surface. He found that little RDX decomposed in the condensed phase. Most recently, Liao and Yang⁷ modeled RDX combustion, utilizing both the gas-phase reaction schemes of Melius and that of Yetter, et al.⁸, which consists of 38 species and 178 reactions. Three condensed-phase reactions were considered along with evaporation of RDX. Bubbles in the liquid layer were treated using a variable porosity concept which allowed retention of the one-dimensional formulation. This model explicitly considers solid, liquid, and gas phases. It was found that up to half of the RDX decomposes in the subsurface two-phase zone and that the gas bubbles occupy 45% of the volume at the surface.

For those models that find little nitramine decomposition in the condensed phase, the

mechanism of surface regression is primarily evaporation. For RDX this is true in three of the four models mentioned previously. This evaporation process, therefore, warrants close scrutiny. In the Ben-Reuven, et al. model, for example, the products of RDX decomposition in the condensed phase (c-phase) are assumed to be dissolved in the remaining liquid RDX and simply entrained in this liquid as it is convected to the surface. These c-phase products then desorb at a rate which is linearly proportional to the RDX evaporation rate. With the possible exception of the Melius model (evaporation rate computation not described), all of these models make use of the equilibrium vapor pressure of pure RDX as a function of temperature. In every case they assume that the heat of vaporization of RDX is not changed by the presence of the c-phase decomposition products. Such an assumption may ultimately prove to be a good approximation where little RDX has decomposed in the c-phase, however, in general it is not. These models also assume that the sticking coefficient, i.e., the fraction of molecules impinging on the surface that actually are absorbed into the surface, for vapor-phase RDX is unity. A value of unity is in fact quite a good assumption for thermal energy vapors; however, in all of the previous treatments, gas-phase species other than RDX are implicitly assumed to have zero sticking coefficients (i.e., only RDX molecules may reenter the surface, despite the fact that the randomly directed thermal velocities are orders of magnitude larger than their convection velocity away from the surface). If one were to allow the non-RDX molecules to reenter the c-phase, it would be necessary to consider molecular diffusion in the liquid layer since their gradient would be steep at the surface and therefore diffusion could well affect the surface concentration. None of the models to date have considered it important to include liquid-phase diffusion.

The subtleties just described could be important since they affect the very mechanism by which the surface regresses. Yet the models discussed above for nitramine combustion are already formidably complex. Drawing firm conclusions about the importance of certain mechanisms using these models is greatly hampered by the considerable uncertainties associated with both the supportive data and physical/chemical mechanisms not being tested. Furthermore, sensitivity analysis cannot entirely relieve the dilemma because it is based on the assumption of small perturbations, and our understanding of many facets of the problem is still rudimentary at this

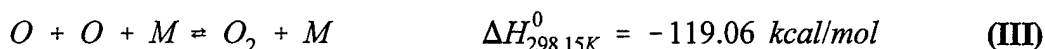
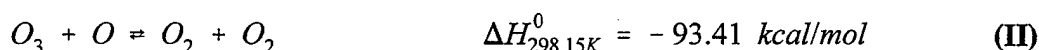
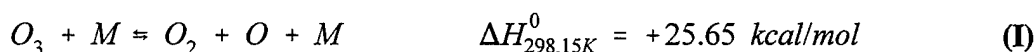
stage. What is needed is a prototypical system that possesses key features of the nitramine problem but with less pervasive uncertainties in supporting data and a less distracting degree of chemical complexity. In our opinion, a study of the steady deflagration of frozen and liquid ozone meets these criteria, and this report describes our current state of progress toward analyzing that problem. It should be admitted, however, that while the frozen ozone system is clearly the best prototypical system from a heuristic viewpoint, it is probably not the most convenient system to adopt from an experimental viewpoint. The sensitivity of the condensed phases of ozone to detonation is legendary and one must work at temperatures below 100 K. After exploiting the ozone case conceptually, it would be wise to seek another prototype of intermediate complexity enabling more intensive comparisons of model to experiment.

2. OZONE PROPERTIES AND DECOMPOSITION CHEMISTRY

In the 1950s liquid ozone was considered as a rocket-propellant ingredient and, consequently, much of the work to characterize its properties, production, and behavior was done during those years. At 1 atm ozone freezes at about 80 K and boils at 161.3 K. The color of gaseous ozone is light blue, liquid ozone indigo blue, and solid ozone deep blue-violet⁹. In any phase O_3 will readily detonate if subjected to heat, spark, flame, or shock. Liquid ozone is particularly sensitive to detonation because rapid heating or even cooling can cause initiation. A common cause of hypersensitivity was found to be certain organic impurities, and eventually it was learned how to avoid such contamination with a consequent increase in safety as well as reproducibility of measurements¹⁰. For example, it was found that suitably purified 100% gaseous ozone detonates reproducibly when its temperature reaches 105°C, whereas contaminated samples had detonated at much lower and unpredictable temperatures. The rate of thermal decomposition of liquid ozone appears not to have been measured, but one can infer that it must be quite slow at temperatures up to 243 K since it was determined not to have affected the measurement of vapor pressure up to that temperature¹¹. Thus, in a model of frozen or liquid ozone combustion, one can safely assume that the effects of any c-phase decomposition are negligible on the time

scale pertinent to a steady deflagration wave. This eliminates a large source of uncertainty in the ozone case relative to the nitramine case. In addition, unlike RDX, the specific heat¹², the thermal conductivity¹³, and the mass density⁹ have all been measured for the liquid phase of ozone. The vapor pressure of ozone has been measured by a number of researchers^{11,14,15} with consistent results.¹⁶ The critical temperature for ozone was measured as 261 K corresponding to a deduced critical pressure of 54.6 atm.⁹ Thermodynamic data and transport data used in the calculations reported in this paper are given in detail elsewhere.¹⁶

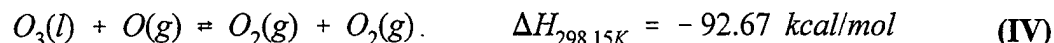
The reaction mechanism for the decomposition of gaseous ozone has been determined with considerable confidence¹⁷ and consists of the following three reversible elementary reactions.



The rate coefficients for the forward directions are taken from Heimerl and Coffee¹⁸ who chose ozone as their prototype of an unbounded gas-phase flame before undertaking flames with more complex chemistry. Rate coefficients for the reverse reactions are computed from the equilibrium constants. These authors did a thorough review¹⁹ of the literature on Reaction I and developed a new expression for the rate coefficient which best represents the experimental rate data over the temperature range 300-3000 K. Because we intended to use this rate at temperatures down to about 160 K, we compared their rate coefficient to the available cold temperature data¹⁷. The fit is quite good in the very low temperature range as well.¹⁶ This single two-parameter Arrhenius rate coefficient accurately represents the rate data over 25 orders of magnitude! Since we intended to do calculations at pressures considerably above 1 atm, we modified the rate coefficient of Heimerl and Coffee¹⁸ for Reaction I to account for the transition of the three-body reaction to a two-body reaction at high pressures. In the absence of more detailed information we used a

Lindemann functionality to describe this pressure saturation effect. The limiting two-body rate was taken from Popovich, et al.²⁰. The rates as used in the calculations reported here in the form required by the PREMIX code are given in Miller¹⁶.

In addition to the three homogeneous gas-phase reactions just discussed, we included the following probable gas/surface reaction.



This equation represents a gas-phase oxygen atom recombining with a liquid-phase ozone molecule on the surface to produce two gas-phase oxygen molecules. This heterogeneous reaction has a large exothermic heat of reaction that is deposited directly on the surface. Reactions II and III are also highly exothermic but heat from those reactions must be conducted from their distributed sites of deposition upstream to the surface. Since the O atom concentration in the gas at the surface is computed in the code, we use the flux of these atoms at the surface temperature multiplied by a reaction probability to compute the reactive flux associated with Reaction IV. Implementation of this new type of reaction requires a modification of the usual boundary conditions in the gas phase at the surface and will be discussed in the next section. Data on this reaction is not yet available but its effects can be bracketed by using the limiting values of 0 and 1 for the reaction probability. This appears to be the first time that the effects of a heterogeneous reaction on the burning rate of an energetic material has been calculated using a detailed chemistry code.

3. DESCRIPTION OF THE CODE

The model developed here considers each phase as a separate mathematical domain in which the mass, energy, and species conservation equations are solved subject to the appropriate boundary conditions conserving the various fluxes across each interface. Pressure changes are considered negligible in this and all of the models of this type, as is molecular diffusion in the liquid phase. The boundary conditions for the case without heterogeneous reaction are fairly

standard and have been discussed elsewhere³. The presence of Reaction IV affects the boundary conditions on the fluxes of species and energy at the gas/liquid interface and the evaporative surface regression mechanism discussed next. The modified species boundary conditions are expressed as follows, P_{hrxn} being the heterogeneous-reaction probability (here taken to be either 0 or 1):

$$Y_O^{-0} \dot{m} = Y_O^{+0} \dot{m} + \rho(YV)_O^{+0} + \frac{1}{4} P_{hrxn} (\rho_O \bar{v}_O)^{+0} \quad (1)$$

$$Y_{O_2}^{-0} \dot{m} = Y_{O_2}^{+0} \dot{m} + \rho(YV)_{O_2}^{+0} - \frac{1}{2} \frac{W_{O_2}}{W_O} P_{hrxn} (\rho_O \bar{v}_O)^{+0} \quad (2)$$

$$Y_{O_3}^{-0} \dot{m} = Y_{O_3}^{+0} \dot{m} + \rho(YV)_{O_3}^{+0} + \frac{1}{4} \frac{W_{O_3}}{W_O} P_{hrxn} (\rho_O \bar{v}_O)^{+0} \quad (3)$$

where \dot{m} is the total mass flux, Y_i^{-0} and Y_i^{+0} are the mass fractions of species i on the liquid and gas sides of the surface, respectively, $(YV)_i^{+0}$ is the diffusion flux of the i th species on the gas side of the surface, and $(\rho_O \bar{v}_O)^{+0}$ is the product of the mass density of atomic oxygen and the average molecular speed of atomic oxygen evaluated at the surface.

The boundary condition on the energy flux at the gas/liquid boundary is modified by the heterogeneous reaction as follows:

$$\begin{aligned} \lambda \left(\frac{dT}{dx} \right)^{+0} = & \dot{m} \left(h_{O_3}^{liq}(T_s) - h_{O_3}^{c-phase}(T_0) \right) + \left(\dot{m} - \frac{1}{4} P_{hrxn} (\rho_O \bar{v}_O)^{+0} \right) \Delta h_{O_3}^{vap}(T_s) \\ & + \frac{1}{4 W_O} P_{hrxn} (\rho_O \bar{v}_O)^{+0} \Delta H_{hrxn}(T_s) \end{aligned} \quad (4)$$

where the specific enthalpy of vaporization at T_s is

$$\Delta h_{O_3}^{vap}(T_s) = h_{O_3}^{gas}(T_s) - h_{O_3}^{liq}(T_s) \quad (5)$$

and the molar reaction enthalpy at T_s for the heterogeneous reaction (Reaction IV) is given by

$$\Delta H_{hrxn}(T_s) = 2H_{O_2}^{gas}(T_s) - H_{O_3}^{liq}(T_s) - H_O^{gas}(T_s) \quad (6)$$

In addition to the heterogeneous reaction at the gas/liquid interface, the model considers evaporation of the energetic material. If there were no reactions, liquid ozone would exist at the surface in equilibrium with its vapor, and the gross evaporation flux would just equal the gross condensing flux. Gas-phase reactions serve to deplete the concentration of vapor molecules and therefore diminish the condensing flux. The evaporating flux is undiminished so that under these conditions a net evaporation flux leads to a regressing surface. One can infer the magnitude of the gross evaporation flux by using the equilibrium value of the condensing flux, which can be calculated from the equilibrium vapor pressure at the surface temperature and the kinetic theory result that the molecular flux crossing a plane in one direction is simply one fourth the product of the number density and the average molecular speed. Under reactive conditions one can use the computed gas-phase density of ozone at the surface to determine the condensing flux. Thus the expression for the net evaporating mass flux of O_3 is

$$\dot{m}_{O_3} = \frac{\alpha W_{O_3}}{4RT_s} \left(\frac{8RT_s}{\pi W_{O_3}} \right)^{\frac{1}{2}} \left(X_{O_3}^{-0} p_{O_3}^e - X_{O_3}^{+0} p_{total} \right) + \frac{W_{O_3}}{4W_O} P_{hrxn} (\rho_O \bar{v}_O)^{+0} \quad (7)$$

where α is the sticking coefficient of the condensing molecules, W_{O_3} is the molecular weight of ozone, T_s is the surface temperature, $p_{O_3}^e$ the equilibrium vapor pressure at T_s , X^{-0} is the mole fraction of O_3 on the liquid side of the surface, X^{+0} is the computed mole fraction of gas-phase ozone at the surface, and p_{total} is the total pressure. The sticking coefficient is expected to be unity

for thermalized vapors, and this value is used exclusively in the calculations reported here. The equilibrium vapor-pressure expression of Jenkins and Birdsall¹¹ is used in all calculations.

If there are liquid components other than O_3 (i.e., for $X^0 < 1$), then the total mass flux can be determined by the following artifice used by Ben-Reuven, et al.³

$$\dot{m} = \dot{m} \left(1 - Y_{O_3}^{-0} \right) + \dot{m}_{O_3} \quad (8)$$

$$\dot{m} = \frac{\dot{m}_{O_3}}{Y_{O_3}^{-0}} \quad (9)$$

where Y^{-0} is the mass fraction on the liquid side of the surface. These relations assume that the components other than O_3 escape the surface according to the same dynamics as O_3 (i.e., these other components are simply entrained by the O_3).

The solutions for each phase are coupled through these boundary conditions, and an iterative approach is used here, solving the equations in each phase for a trial set of boundary conditions that are then adjusted and new solutions obtained in each domain until convergence is judged to have been achieved. The mass flux \dot{m} , surface temperature T_s , and the liquid-layer thickness X_{liq} are the eigenvalues of the problem, and values are sought that satisfy the heat flux boundary conditions at the solid/liquid and liquid/gas interfaces and the evaporation mechanism constraint. In the gas phase the solution is obtained using the PREMIX code²¹. Though for the ozone case we consider no chemical reactions in the liquid phase, in anticipation of applying the code to other energetic materials, we have created a special version of PREMIX to treat the liquid phase. Therefore, our code is capable of treating an arbitrary number of gas and liquid reactions. Although the same strategy could be followed for the solid phase, at present, there is no capability for reactions there; however, a numerical integration over the solid phase is utilized permitting temperature-dependent properties there. The calculations reported here include the effects of thermal diffusion, though the burning rate changes by only a few percent if it is not considered. Also, the multicomponent transport properties were evaluated and used in the gas phase.

Numerical accuracy of the burning rate and heat feedback was examined using up to 15,000 grid points in the gas phase. Results presented here used about 4,500 grid points in the gas phase and should ensure accuracy of a few tenths of a percent in the computed burning rates and heat feedback.

4. BURNING RATES AND THERMAL STRUCTURE

The computed burning rate for frozen ozone at an initial temperature of 40 K as a function of pressure is given in Fig. 1 and Tables 1 and 2. The first thing to notice is the relatively high value of the burning rate, about 2 cm/s at 10 atm. At that pressure and an initial temperature of 300 K, RDX burns an order of magnitude more slowly! The pressure dependence is also well approximated by the power-law dependence characteristic of many energetic materials; the pressure exponent is slightly less than 1, similar to RDX.

Fig. 2 shows the temperature and species profiles in each phase at 1 atm. On the scale of

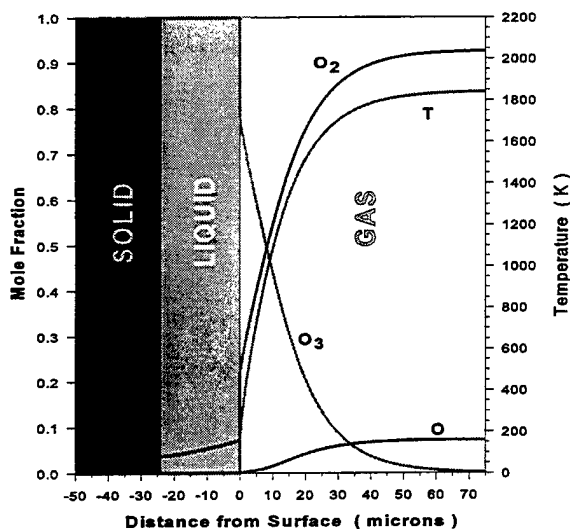


Figure 1. Computed temperature and species profiles in three phases for deflagrating frozen ozone at 1 atm and an initial temperature of 40 K.

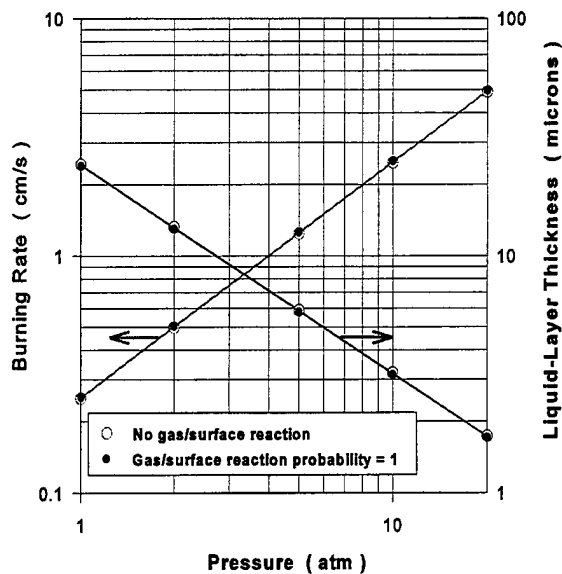


Figure 2. Computed burning rates and liquid-layer thickness vs. pressure for frozen ozone at 40 K assuming maximum and minimum effects of gas/surface reaction.

this figure the results with maximum and zero heterogeneous reaction are indistinguishable, and the major difference in results between the two assumptions is in the O-atom concentration at the surface, being about 30 times smaller for the maximized effect of Reaction IV. Tables 1 and 2 summarize the numerical results.

TABLE 1. Summary of Results for 100% Solid Ozone without Gas/Surface Reaction at 40 K Initial Temperature. T_{bp} is the boiling point temperature.

p (atm)	r (cm/s)	T_s (K)	X_{liq} (microns)	$\lambda(dT/dx)^{+0}$ (cal/cm ² -s)	T_{bp} (K)
1	0.2497	157.9	24.32	59.77	161.3
2	0.4985	168.4	13.22	120.8	172.3
5	1.242	184.8	5.89	307.0	189.6
10	2.470	199.6	3.20	621.7	205.5
20	4.906	217.4	1.73	1262.	224.7

TABLE 2. Summary of Results for 100% Solid Ozone with Unit Probability for Gas/Surface Reaction at 40 K. Φ_{hrxn} is the last term in Eqn. (8).

p (atm)	r (cm/s)	T_s (K)	X_{liq} (microns)	$\lambda(dT/dx)^{+0}$ (cal/cm ² -s)	Φ_{hrxn} (cal/cm ² -s)
1	0.2526	158.1	24.07	51.39	8.73
2	0.5052	168.5	13.06	104.2	17.6
5	1.257	184.9	5.83	264.8	44.4
10	2.505	199.8	3.16	538.5	89.2
20	4.953	217.6	1.72	1090.	179.

We were able to find only one experimental value of the burning rate of condensed-phase ozone. Streng²² measured a value of 0.4 cm/s for a liquid mixture of 90% ozone with 10% molecular oxygen at an initial temperature of 90 K. Our calculations predict 0.27 cm/s at these conditions. While the computed value is in rough agreement with the experimental value, we believe that the quality of our input data warrants closer agreement. Of course, the experimental value may be inaccurate; the liquid was contained in a 9 mm Pyrex tube cooled outside by liquid oxygen and therefore the flame could have led to preheating of the liquid by conduction along the tube walls. However, our calculations show that an initial temperature of even 160 K is not enough to boost the rate to 0.4 cm/s. There is also the possibility of hydrodynamic instabilities in the liquid surface; this also would give a falsely high value to the measured rate. No visual observations of this sort were noted by the authors, who generally reported their experiments with considerable care. On the other hand, the boiling point of O₂ is very much less than that of O₃ (90 K compared to 161 K); therefore, the O₂ component should lower the boiling point of O₃ in the mixture and decrease its heat of vaporization. We may be seeing a case here where the heat of vaporization is significantly affected by the presence of as little as 10% other molecules. This issue may have direct relevance to the RDX case. (See discussion in Section 6.)

5. MECHANISM ANALYSIS

Fig. 1 shows that the maximum effect of the gas/surface reaction is slight, enhancing the rate by about 1% at 1 atm and 20 atm. Calculations have not been carried out at pressures greater than 20 atm because the model would have to be modified to account for the approach to the thermodynamic critical point at about 55 atm. There are two mechanisms by which the heterogeneous reaction might bolster the rate. Its heat of reaction is deposited directly at the surface, whereas the heat from gas-phase reactions must be conducted upstream; therefore, the heterogeneous reaction is more efficient in having its heat affect the burning rate. The other mechanism is the destruction of liquid-phase ozone by oxygen atoms, a process which contributes

directly to the regression rate. Analysis of the calculations show that most of the heterogeneous-reaction contribution to the burn rate is by this latter mechanism. To understand how the heterogeneous reaction affects the gas-phase reaction mechanism, it will be helpful to examine the role of the various reactions in different parts of the flame.

Fig. 3 shows how the net rate of production of each species is spatially distributed through the flame. Four spatial zones are identified in the figure. In Zone A, adjacent to the surface, O_3 is being produced at the expense of O_2 and O . The region of most intense reaction activity is divided into two zones, B and C. In Zone B O_3 is now being destroyed, O_2 is now being produced, and O is being destroyed more vigorously than at the surface. In Zone C, O_3 continues to be destroyed and O_2 continues to be formed, but now O is being produced. Zone C is the site of most of the O_3 destruction. Finally, by Zone D, O_3 has been consumed and, though the scale of this figure does not show it, O_2 is being produced slowly and O is being destroyed slowly.

The mechanism is more completely revealed by examining the contribution to each species production rate by each reaction. An examination of these rates shows that immediately adjacent to the surface in Zone A, Reaction I is dominating and it is running in reverse! This is enabled by the diffusion of O atoms upstream from

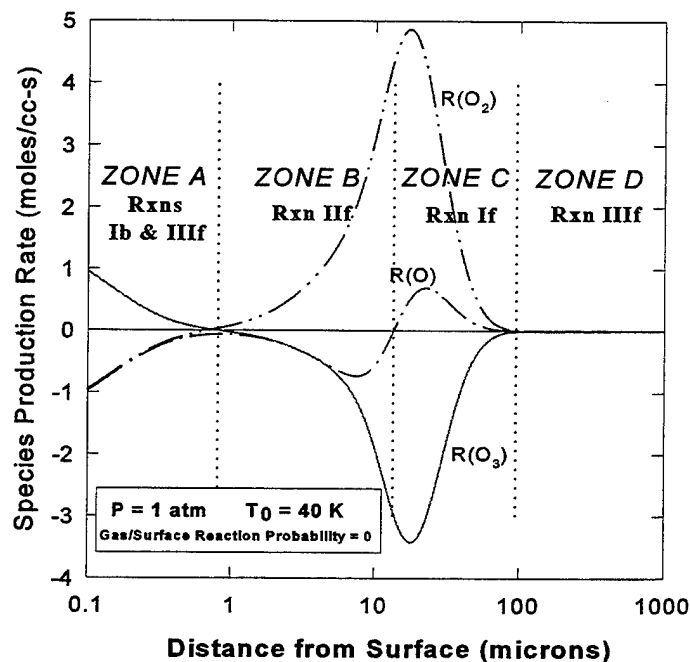


Figure 3. Net species production rates in the gas phase of deflagrating frozen ozone at 1 atm and 40 K without heterogeneous reaction.

where they are produced in Zone C. Note that this Zone-A reaction is exothermic. Reaction III, in the forward direction, also contributes here. In Zone B Reaction II dominates, in the forward direction, producing O_2 at the expense of O_3 and O . Zone C is dominated by Reaction II and Reaction I, this time in the forward direction, producing O_2 and O at the expense of O_3 . Finally, the recombination Reaction III dominates in the post-flame region, Zone D.

An unexpected outcome of this mechanism analysis is that **the first chemical step (spatially) in the steady deflagration of frozen ozone is the production of ozone!** This unusual result had never been noted previously in studies of ozone gas flames¹⁸, so we did an unbounded flame calculation for gas-phase ozone at 161 K. A small but distinct peak in the total heat-release profile due to Reaction I running in reverse could be seen in the leading edge of the flame consistent with the three-phase calculations. When the free flame was given an initial temperature of 300 K (where previous studies had been conducted), this stage disappeared completely.

Returning to the three-phase case, the effect of the reactions on the heat release profiles at 1 atm is shown in Fig. 4. It can be seen that the "simple" but real ozone chemistry manifests three distinct gas-phase stages of heat release. The stage closest to the surface results from the recombination of O and O_2 to form O_3 , Reaction I running in reverse and the forward direction of Reaction III. This first stage coincides with Zone A. The next stage comprises Zones B and C. As can be seen in the figures, only Zones A and B are affected by the heterogeneous reaction which robs both zones of its O reactant. The final heat release stage is Zone D where O atoms slowly recombine

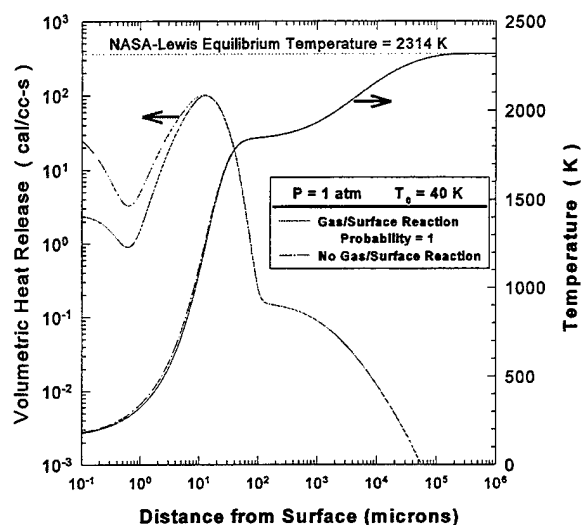


Figure 4. Gas-phase heat release and temperature profiles showing effect of heterogeneous reaction at 1 atm. Both Zones A and B of Fig. 3 are affected.

to form the final product O_2 .

The reason the heterogeneous reaction has such a slight effect on the burning rate can now be understood. First, note that the heat feedback to the surface can be expressed in terms of the volumetric heat release from the gas-phase reactions, $q(x)$, by the following simple expression (valid for constant c_p and λ)

$$\lambda \left(\frac{dT}{dx} \right)_{+0} = \int_0^{\infty} q(x) e^{-\frac{\dot{m}c_p x}{\lambda}} dx. \quad (10)$$

This expression shows that the heat released within a distance of about $\lambda/\dot{m}c_p$ is returned to the surface with good efficiency. At 1 atm this characteristic distance is of the order of 10 microns. By Fig. 4 one can see that the heterogeneous reaction decreases the gas-phase heat release within this distance. Thus heat gained at the surface from the heterogeneous reaction would have come back to the surface conductively from the gas phase anyway. The net effect is therefore almost neutral.

6. FUTURE WORK

A number of questions remain on the ozone problem that have an important bearing on the modeling of other energetic materials. As alluded to in the Introduction, there is an inconsistency in all of the existing models (including the one in this paper) in the fact that the vapor-phase energetic material (ozone, RDX, HMX, etc.) is assumed to have unit sticking coefficient whereas the other gas-phase species are assumed to have a zero value. Relaxing this unphysical assumption (i.e., the zero values) will require consideration of molecular diffusion in the liquid layer. While it is true that diffusion in the liquid phase is generally much slower than in the gas phase, the importance of the process depends on the magnitude of the concentration gradients and these are expected to be very large close to the surface.

Secondly, the evaporation process has been highly idealized in existing models (again including the present one). Both the heat of vaporization and the rate of escape from the liquid surface has been assumed not to depend on the presence of other liquid-phase molecules. These other liquid-phase molecules could be either decomposition products of liquid-phase reactions or gases adsorbed due to a nonzero sticking coefficient. What is needed to explore these effects is a broadly applicable theory for predicting heats of desorption from multicomponent liquids and a kinetic theory of evaporation from multicomponent liquids. The current artifice of using the measured vapor pressure to estimate the gross rate of evaporation should be valid for a pure substance, but of little value for a multicomponent liquid, where both the heat of desorption and the rate of escape from the surface will be affected by the presence of other molecules.

Thirdly, extending model validity to pressures through the thermodynamic critical point may have considerable importance to the combustion of liquid energetic materials. This extension is related to the ability to predict heats of desorption from multicomponent liquids, as such a theory must account for the heat of desorption (or vaporization for a pure substance) going to zero at the critical point. Although beyond the scope of this paper, these areas are now being addressed and progress will be described in a future report.

We hope that through this report and the outline given of future work, that a compelling case has been made for the value of modeling prototypical systems. While frozen or liquid ozone is the obvious choice from the standpoint of theoretical convenience, from an experimental viewpoint it is an extremely hazardous and inconvenient material to work with due to its detonation sensitivity and low melting/boiling points. Hydrazine (N_2H_4), with melting and boiling points near those of water, may be a natural compromise for a combined experimental and theoretical effort. We plan to explore further the feasibility of studying that system.

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4. TITLE AND SUBTITLE Three-Phase Combustion Modeling: Frozen Ozone, a Prototype System			5. FUNDING NUMBERS PR: 1L161102AH43	
6. AUTHOR(S) Martin S. Miller				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) U.S. Army Research Laboratory ATTN: AMSRL-WM-PC Aberdeen Proving Ground, MD 21005-5066			8. PERFORMING ORGANIZATION REPORT NUMBER ARL-TR-1320	
9. SPONSORING/MONITORING AGENCY NAMES(S) AND ADDRESS(ES)			10. SPONSORING/MONITORING AGENCY REPORT NUMBER	
11. SUPPLEMENTARY NOTES This report was an invited paper at the Materials Research Society Fall Meeting, 27-29 Nov 95, Boston, MA, and was published as a part of the proceedings of that meeting.				
12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.			12b. DISTRIBUTION CODE	
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14. SUBJECT TERMS combustion, model, ozone, RDX			15. NUMBER OF PAGES 28	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL	

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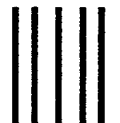
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